Aerosol processing by clouds: Observations from the HCCT-2010 hill cap cloud experiment

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HCCT-2010

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OBJECTIVE AND EXPERIMENTAL

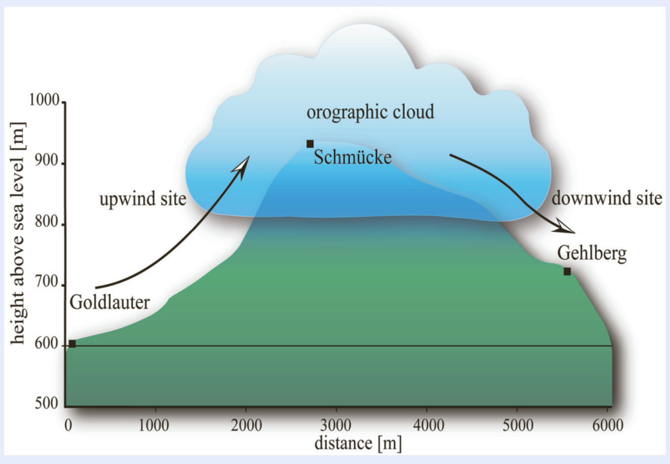

Fig. 1: Scheme of the campaign area

- Hill Cap Cloud Thuringia 2010 (HCCT-2010): Ground-based cloud experiment on physical and chemical aerosol-cloud interaction at Mt. Schmücke, Germany, in Sep and Oct 2010
- Lagrangian-type approach with three sampling sites (Fig. 1): upwind, in-cloud, downwind
- Berner impactors at upwind and downwind sites for offline chemical characterization
- Four aerosol mass spectrometers (AMS, Aerodyne): 1 upwind, 2 in-cloud , 1 downwind
- Interstitial Inlet (INT) for sampling of interstitial particles and gas phase, CVI inlet for sampling cloud droplet residuals (Mertes et al., 2005)
- Analysis of impactor samples for inorganic ions (IC), OC/EC (thermographic method), WSOC (TOC-analyzer), dicarboxylic acids (CE)
- Comprehensive analysis of meteorology and local air flow conditions during campaign (Tilgner et al., 2013)

→ Several "Full Cloud Events" (FCE) and "Non-Cloud Events" (NCE) with connected airflow at all sites and overall suitable conditions for Lagrange-type data analysis approach (Tables 1)

Table 1: Event list

			Impactor sampling times	
event	**start CEST**	**stop CEST**	**start CEST**	**stop CEST**
FCE1.1	14/09/2010 11:00	15/09/2010 01:50	14/09/2010 11:00	15/09/2010 02:00
FCE1.2	15/09/2010 03:00	15/09/2010 06:20		
FCE2.1	15/09/2010 23:00	16/09/2010 02:00		
FCE4.1	16/09/2010 13:10	16/09/2010 15:00		
FCE5.1	16/09/2010 21:40	16/09/2010 23:50		
FCE7.1	24/09/2010 21:10	25/09/2010 00:50	24/09/2010 23:45	25/09/2010 01:45
FCE11.2	01/10/2010 20:50	02/10/2010 03:10	01/10/2010 22:30	02/10/2010 05:30
FCE11.3	02/10/2010 07:10	03/10/2010 00:30	02/10/2010 14:30	02/10/2010 20:00
FCE13.3	06/10/2010 06:50	07/10/2010 01:00	06/10/2010 12:15	07/10/2010 03:15
FCE22.0	19/10/2010 01:50	19/10/2010 09:00		
FCE22.1	19/10/2010 21:10	20/10/2010 02:30	19/10/2010 21:30	20/10/2010 03:30
FCE24.0	21/10/2010 22:10	22/10/2010 10:00		
FCE26.1	23/10/2010 23:40	24/10/2010 07:20	24/10/2010 01:30	24/10/2010 08:45
FCE26.2	24/10/2010 08:40	24/10/2010 12:20	24/10/2010 09:15	24/10/2010 11:45

			Impactor sampling times	
SW events	**start CEST**	**stop CEST**	**start CEST**	**stop CEST**
NCE0.1	15/09/2010 12:10	15/09/2010 22:10	15/09/2010 18:00	15/09/2010 23:30
NCE0.2	16/09/2010 02:50	16/09/2010 06:10		
NCE0.3	19/09/2010 11:20	19/09/2010 17:00		
NCE0.4	20/09/2010 11:50	20/09/2010 16:20	20/09/2010 10:30	20/09/2010 20:30
NCE0.5	22/09/2010 10:50	22/09/2010 16:00		
NCE0.6	23/09/2010 23:30	24/09/2010 02:50		
NCE0.7	24/09/2010 10:30	24/09/2010 20:00		
NCE0.8	03/10/2010 11:20	03/10/2010 14:40		
NCE0.9	21/10/2010 12:40	21/10/2010 21:20	21/10/2010 14:20	21/10/2010 22:10
NCE0.10	23/10/2010 16:10	23/10/2010 22:40		
NE events				
NE_NCE0.1	07/10/2010 13:00	07/10/2010 18:50		
NE_NCE0.2	08/10/2010 15:10	08/10/2010 18:30		
NE_NCE0.3	09/10/2010 14:30	10/10/2010 09:30		
NE_NCE0.4	10/10/2010 15:50	11/10/2010 03:30		
NE_NCE0.5	11/10/2010 13:00	12/10/2010 04:40		

RESULTS AND DISCUSSION

AMS concentrations at all sites for three cloud events

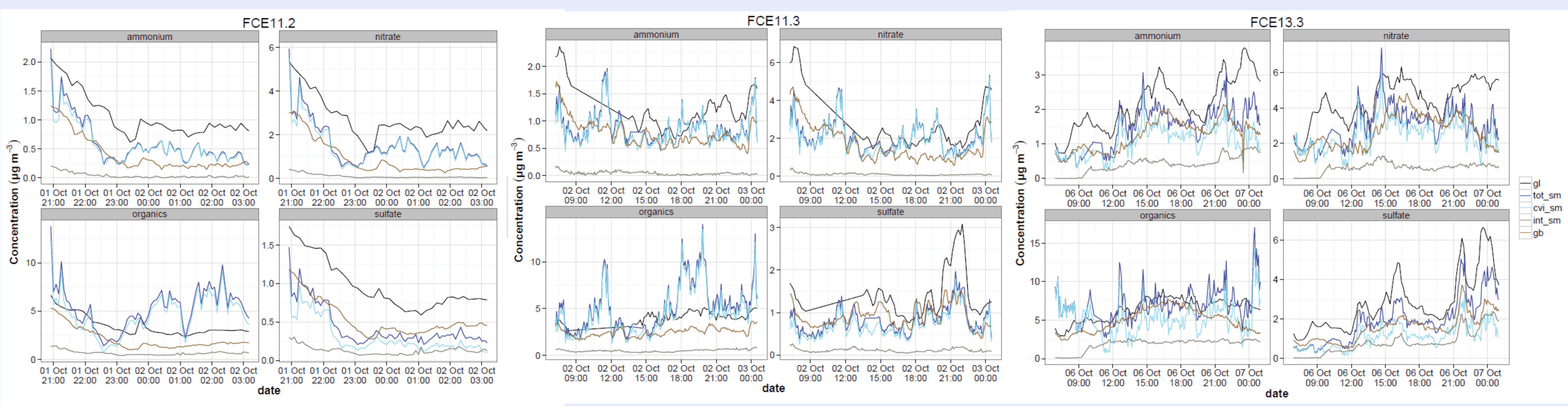

Fig. 2: AMS concentrations at 3 sites: upwind (Goldlauter, gl), in-cloud interstitial (Schmücke, int_sm), in-cloud droplet residuals (cvi_sm), in-cloud total (int+cvi, tot_sm), downwind (Gehlberg, gb).

- Intercomparison of AMS concentrations with Berner impactor PM1.2 data showed larger discrepancies at downwind site, especially for ammonium and nitrate (factor 1.8 different on average, not shown)

→ To ensure comparability of AMS datasets, the impactor measurements were used as reference and the AMS downwind data was scaled to have same mean Berner/AMS ratio as at upwind site

- Fig. 2 shows time series of main AMS compounds for three longer FCEs (11.2, 11.3, 13.3)

→ Usually upwind conc. > downwind conc., in-cloud conc. often somewhere in between

Concentrations upwind – downwind for all events

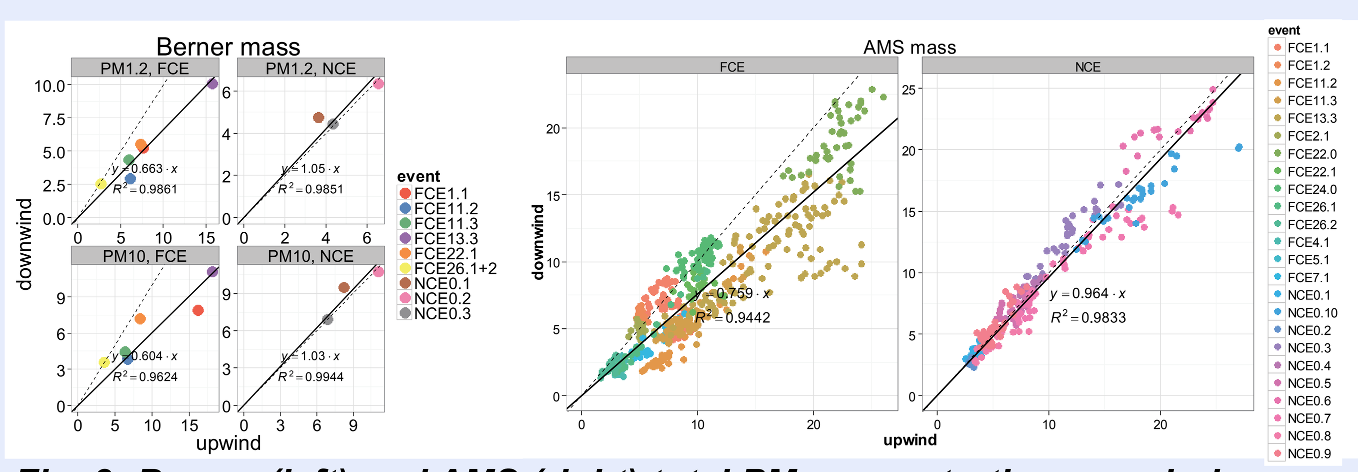

Fig. 3: Berner (left) and AMS (right) total PM concentrations upwind versus downwind for all FCEs and NCEs. All data in µg m^{-3}.

→ Downwind concentration decrease seen only during FCEs (Fig. 3). During reference NCEs (connected air flow, no hill cap cloud) concentrations at the two sites agree reasonably well.

→ Physical loss processes during cloud events, e.g. by droplet scavenging by trees and/or entrainment of cleaner air masses from aloft

AMS mass fractions upwind – downwind for three cloud events

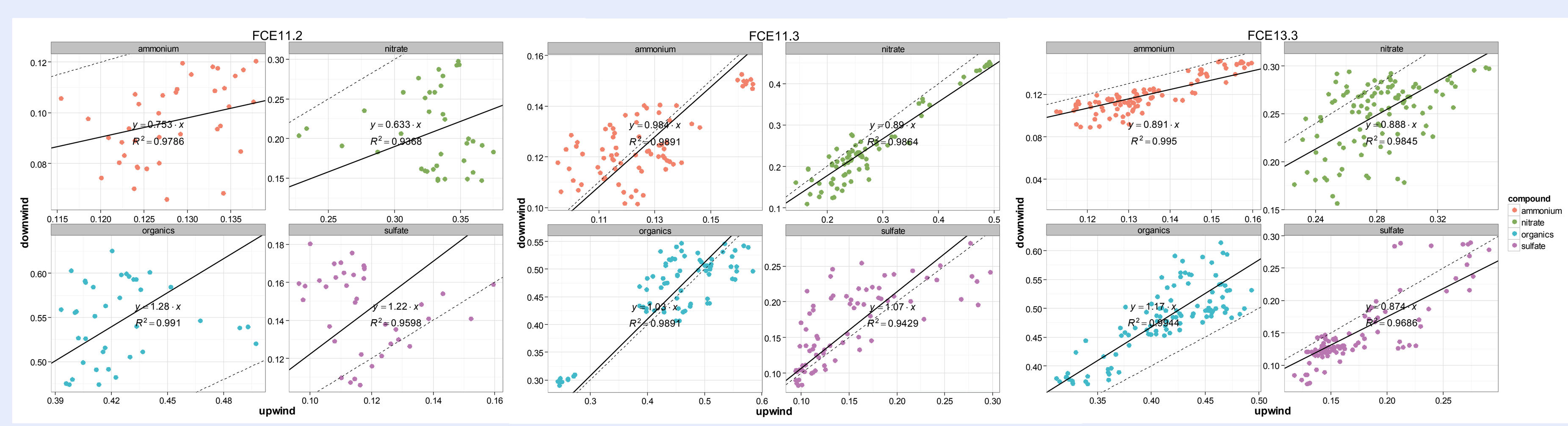

Fig. 4: AMS mass fractions (fraction of compound to total AMS mass upwind versus downwind

- Mass fractions can indicate chemical modifications on a relative scale
- Fig. 3 shows scatter plots of main AMS components mass fractions upwind versus downwind for three longer FCEs (11.2, 11.3, 13.3)

→ sometimes only slight changes (FCE11.3), sometimes sulfate and/or organics strongly increased downwind (FCE11.2, FCE13.3)

Berner and AMS downwind mass addition during all cloud events scaled by different „conservative" tracers

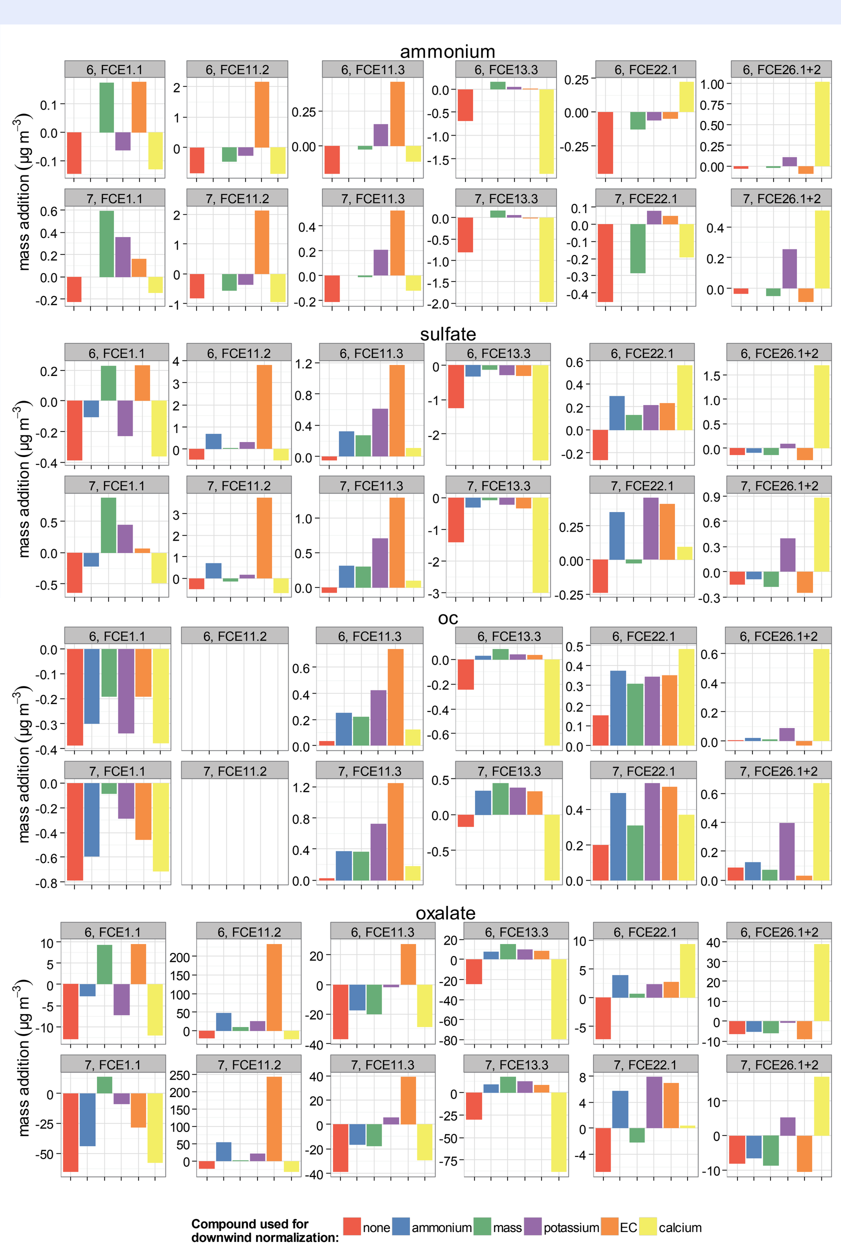

Fig. 5: Berner downwind mass additions ($c_{downw.}$ - $c_{upw.}$) for measured data (red) and data scaled using different „conservative" tracers measured from the same impactor samples.

- Scaling the downwind site by a conservative tracer (chemically inert, physical losses similar to compound of interest) can reveal whether cloud passage has added mass to pre-existing (upwind) particles
- As the ideal conservative tracer is not available, several ones were used for impactor (NH_4^+, mass, K^+, EC, Ca^{2+}) and AMS scaling (NH_4^+, number conc. of small particles with D_p=25-49 nm, BC)
- Fig. 5 shows results for some compounds from impactor measurements: All tracers indicate mass addition of sulfate and OC at least during FCE11.3 and FCE22.1in the order of approx. 0.2 - 0.4 µg m^{-3} . Similarly, oxalate increases during FCE11.2 and FCE22.1 in the order of 5 – 50 ng m^{-3}
- AMS data (Fig. 6) usually similar to impactor data. Several events exist where all parameters indicate mass addition.

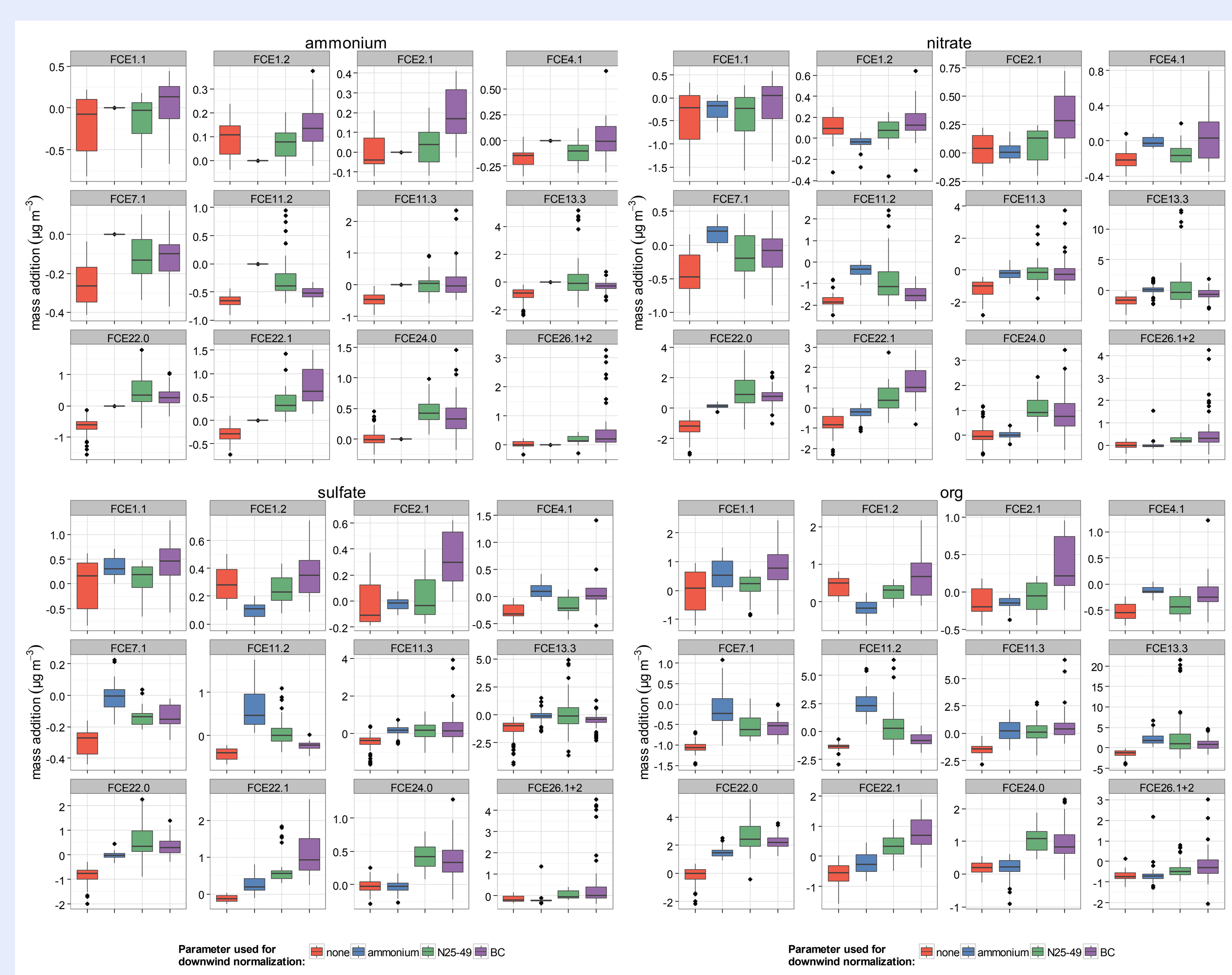

Fig. 6: Box-plots of AMS mass additions ($c_{downw.}$ - $c_{upw.}$) for measured data (red) and data scaled using different continuously measured „conservative" tracers.

CONCLUSIONS

- Hill cap cloud used as a natural flow through reactor during HCCT-2010
- Physical mass losses during cloud events challenge a direct observation of chemical modifications after cloud passage
- Mass fractions indicate modified relative chemical composition
- Using best-available "conservative" tracers to correct for physical losses results in several cloud events where mass increases after cloud passage in the order of several tenths of µg m^{-3} are observed for sulfate, organics and sometimes nitrate.

REFERENCES AND FUNDING

Mertes et al., 2005, Atmos. Environ. 39(23-24), 4233-4245

Tilgner et al., 2013, submitted to ACPD

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